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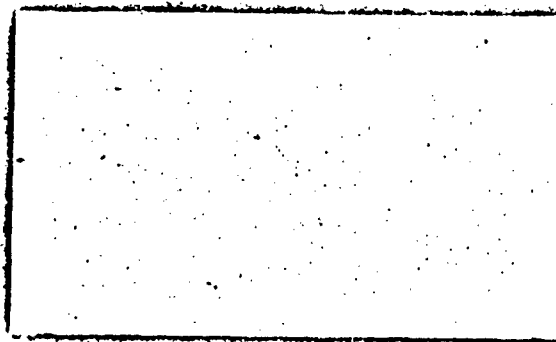
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ELEVENTH MONTHLY PROGRESS REPORT

Covering the Period

March 1 to March 31, 1957

Title: Detection of V-Agents

Prepared by

R. H. Poirier, R. D. Morin, S. J. Kiehl,
R. W. Pfeil, and A. E. Bearse

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ELEVENTH MONTHLY PROGRESS REPORT

on

DETECTION OF V-AGENTS

to

**CHEMICAL CORPS
CHEMICAL WARFARE LABORATORIES**

from

BATTELLE MEMORIAL INSTITUTE

by

**R. H. Poirier, R. D. Morin, S. J. Kiehl,
R. W. Piest, and A. E. Bearse**

March 31, 1957

INTRODUCTION

The development of simple, direct, and sensitive chemical tests for the detection of V-agents in both liquid and aerosol forms is the main objective of this project. Other objectives include the development of methods for the detection of V- and G-agents, or their degradation products, in the presence of decontaminating agents of the active-halogen type.

This is the Eleventh Monthly Progress Report on this project and summarizes the work completed during the period March 1 to March 31, 1957. Included are results on the detection of V-agents by means of Michler's ketone oximes, Dragendorff's reagent, quinone oximes, and halogenated quinones. Further attempts to use acid indicators, namely Congo red, for the detection of V- and G-agents, or their degradation products in the presence of decontaminants are also reported.

A proposed course of study on the detection of V-agents through their thione functions by the use of tetrazolium reagents is outlined.

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SUMMARY

The development of detector papers and crayons based on Michler's ketone oxime (MMK-O) reagent systems was continued. Attempts to use aromatic and aliphatic mercaptans to protect MMK-O and EMK-O* against the deteriorative action of light were unsuccessful. The concentration of zinc thiocyanate in MMK-O crayons greatly influenced the sensitivity of the crayon toward EA-1701. The reagents 2-dimethylaminoacetophenone oxime and the sodium salt of isonitroso-4-dimethylaminoacetophenone did not show much promise as reagents for V-agents, but formed a basis for new oxime reagents.

Investigation of Dragendorff's reagent as a means for detecting V-agents through their amine function included studies on crayon formulations, reagent variations, and analysis of a color body from a modified Dragendorff reagent. Crayons that were prepared from a modified Dragendorff's reagent powder formulated according to ACC directions were tan and left marks on paper that became black upon storing at 65°C. Freshly formed marks gave a distinct orange test color with EA-1701 plus water, but the aged marks developed only a very faint orange color with this agent. Numerous variations were made in the ACC modified Dragendorff system, and the formulation that gave the most encouraging results with regards to activity and stability contained bismuth oxychloride, metaphosphoric acid, calcium chloride, and potassium iodide. This system gave light tan crayons that gave a bright red test color with EA-1701. Marks from these crayons turned orange brown at room temperature with no apparent loss of activity. The marks darkened on storage at 65°C. and after this treatment only gave specks of red color with agent. Analysis of a color body isolated from the reaction of the modified Dragendorff's reagent and tri-n-butylamine indicated that appreciable quantities of chloride are present along with the iodide.

Three analogues of quinonemonoxime (4-nitrosophenol) were examined in the conversion of quinone oximes to intensely colored indophenols by the action of acylating agents and phenols. Apparently, only strong acylating agents, such as benzene sulfonyl chloride, are capable of effecting the interaction of quinone oximes and phenols. Less active acylating agents, such as diethyl ethanephosphonate (DEEP) and EA-1701 are ineffective in this reaction. Metal salts that normally enhance the action of acylating agents upon Michler's ketone oximes did not duplicate this activation in the reactions of quinone oximes with DEEP and EA-1701.

Additional work on the use of Congo red acid indicator for the detection of V- or G-agents, or their degradation products, in the presence of decontaminating agents failed to show promise. Other detection systems are being sought. Presently, tetrazolium salts, particularly 2-(4-iodophenyl)-3-(4-nitrophenyl) tetrazolium chloride and 2-(5-methylthiazol-2-yl)-3,5-diphenyltetrazolium bromide, are being considered.

*Ethyl homologue of Michler's ketone oxime.

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EXPERIMENTAL*

Detection of V-Agents

Michler's Ketone Oximes

Stability of Test Papers and Crayons. Previous attempts to stabilize Michler's ketone oximes by means of antioxidants and free-radical chain terminators have included testing of a wide variety of phenols and aromatic amines as well as certain inorganic salts. However, a patent recently issued to A. L. Lynch** claims that aromatic mercaptans stabilize aromatic amines against atmospheric oxidation. Although the deterioration of MMK-O and EMK-O is believed to result from photo-induced oxidation, nevertheless, it appeared of interest to attempt the use of aromatic and aliphatic mercaptans for the stabilization of Michler's ketone oximes. Preliminary evaluation of this approach was thus made by examining the effect of several widely different mercapto compounds upon the stability of EMK-O test papers to fluorescent light. The compounds that were evaluated are listed below:

2-Naphthalenethiol

2-Mercaptobenzoic acid

1-Mercaptobenzothiazole

2-Mercaptobenzothiazole

2-Mercaptothiazoline

Mercaptosuccinic acid

Thiodiglycolic acid

The test papers were prepared by wetting filter paper discs (Whatman No. 2) in duplicate with an acetone solution containing 1 per cent EMK-O and 0.2 per cent mercapto compound. One set of papers was exposed to fluorescent light***, whereas the other set of papers was stored in the dark for reference. None of the mercapto compounds that were evaluated gave satisfactory stabilization of EMK-O papers. In fact, except for 2-naphthalenethiol, addition of these compounds caused EMK-O papers to turn bright yellow as compared to pale yellow for the control paper upon exposure to light. The naphthalenethiol-EMK-O papers, however, did not discolor any more than the control paper in light, and remained white in the dark as compared to pale yellow for the control. In spite of this apparently superficial improvement, it seemed desirable to evaluate the stabilization effect of 2-naphthalenethiol on crayons before

*The original data are recorded in Battelle Laboratory Record Books Nos. 12872, 12903, and 13023.

**U. S. Patent 2,737,137, July 31, 1956.

***The light source consisted of two 15-watt cool light fluorescent bulbs placed approximately 12 inches from the test papers.

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discontinuing the examination of mercaptans as light stabilizers for Michler's ketone oximes.

The crayons that were formulated for the evaluation of 2-naphthalenethiol as a potential stabilizer were based on MMK-O, zinc thiocyanate, and chalk as previously used in the studies on antioxidants. A detailed description of the preparation of such crayons is presented in the Appendix of this report. Test papers consisting of filter paper streaked with marks from MMK-O crayons containing 2-naphthalenethiol, with and without zinc thiocyanate, appeared to discolor as badly as the controls during exposure to fluorescent light for 24 hours. The thiol decreased perceptibly the activity of the MMK-O crayon streaks toward 1 per cent EA-1701 in benzene.

Only a few of the compounds mentioned by Linch were tried in these experiments, but it is unlikely that a more thorough search of mercapto compounds would solve the light stability problem of Michler's ketone oximes. It now appears that the photosensitivity of MMK-O and related oximes might best be diminished through structural variations within the oxime reagents. This factor is being considered in the current syntheses of Michler's ketone analogues.

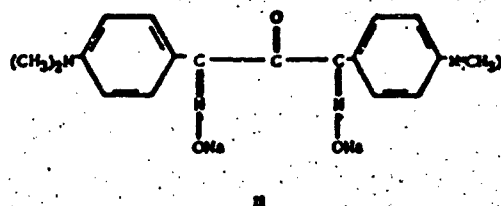
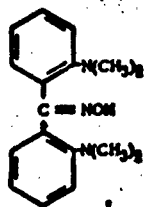
MMK-O Crayon Formulation Studies. Through spectrophotometric studies it was found that the rate of color formation in systems consisting of EMK-O, zinc thiocyanate, and p-toluenesulfonyl chloride is dependent primarily on the concentration of oxime and sulfonyl chloride. Variations in their concentration caused proportional changes in the rate of color formation. However, variations in the concentration of zinc thiocyanate influenced less significantly the color reaction. Although this kinetic study was still in the preliminary stages, it was of interest to establish whether or not some of these relationships held in crayon systems. Hence, crayons that contained MMK-O and zinc thiocyanate in varying molecular ratios were formulated and tested on benzene solutions of EA-1701. The results of these tests seemed to agree with the results of the kinetic studies in that the activity of the crayon showed a greater dependency upon the content of MMK-O than upon the content of zinc thiocyanate. However, zinc thiocyanate was vitally necessary for reactions with EA-1701; no color reaction was obtained in its absence.

New MMK-O Analogues. It was indicated in the Tenth Monthly Progress Report that simple analogues of MMK-O, namely 4-dimethylaminobenzaldehyde oxime (anti) and 4-dimethylaminoacetophenone oxime, did not give striking color reactions with acylating, sulfonating and phosphorylating agents, including EA-1701, even in the presence of zinc thiocyanate or mercuric thiocyanate. However, in an effort to establish a sound basis of approach in the development of new oxime reagents, the simple analogues 2-dimethylaminoacetophenone oxime and the sodium salt of isonitroso-4-dimethylaminoacetophenone were prepared and examined. These model compounds showed little promise as reagents for the detection of G- and V-agents, but their apparent light stability enhanced the interest in the ortho analog of MMK-O (I), and the DIA-MMK-O hybrid (II) that are now being synthesized.

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Dragendorff's Reagent

A sample of modified Dragendorff's reagent powder that was prepared according to the directions of B. Fromm, Army Chemical Center, was pressed into crayons. These crayons were tan colored and left tan marks on paper, which darkened to a deep orange-brown within a few weeks at room temperature, but still gave an orange test color with a 1 per cent solution of EA-1701 in the presence of water. After accelerated aging for 2 weeks at 65°C. both the crayon and marked filter paper had turned black and developed only a very faint orange color with V-agent.

Variations were made in the composition of the modified Dragendorff's reagent in an attempt to improve its stability, test color, and water requirement. By replacing the bismuth oxychloride with an equivalent amount of bismuth in the form of bismuth trichloride a dull gray powder was obtained that gave a black, metallic appearing crayon. Although moistened marks from this crayon became orange when treated with EA-1701, the black background made the orange color difficult to see. In addition, crayon marked papers that were stored for 2 weeks at 65°C. developed only a fleeting orange color. A further variation of the modified Dragendorff's reagent was the use of bismuth triiodide as the source of bismuth. This formulation, too, produced a shiny black crayon that gave difficultly distinguishable tests with EA-1701. Marks from this crayon after storage for 2 weeks at room temperature, or at 65°C., were insensitive to EA-1701, but the addition of acid reactivated the marks. Substitution of sodium chloride for the calcium chloride gave a tan powder and a gray crayon that responded readily to V-agent initially, but sluggishly after 2 weeks at room temperature or 65°C.

The elimination of metaphosphoric acid, and the substitution of bismuth trichloride for bismuth oxychloride led to a light tan powder and a brownish-black crayon. Test papers freshly prepared from such a crayon gave a bright red test color with EA-1701 in the presence of water. However, after 2 weeks' storage at room temperature these papers only gave a speckled red color with V-agent, and after 2 weeks at 65°C. they were inactive.

The use of potassium iodide as the source of iodide, in place of the cadmium iodide that is required in the original modified formulation, gave a white powder and a white crayon. Test papers made from these crayons slowly developed an orange color with solutions of EA-1701 and water. The activity of these papers was unchanged by storage at room temperature for 2 weeks, but was diminished considerably by storage at 65°C., even though the papers did not darken. Since the potassium iodide seemed to provide a stable source of iodide, a formulation with a tenfold increase in the

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amount of potassium iodide was next examined. The powder and the resulting crayon thus obtained were light tan, and formed marks that gave a bright red color with EA-1701 plus water. After 2 weeks at room temperature the test marks had turned orange brown but still gave a bright red color with agent; however, after 2 weeks at 65°C. the marks had darkened and only gave a spotty red color with agent.

None of the above variations of the modified Dragendorff's formulation gave a stable reagent. However, the red test color obtained with the formulation containing potassium iodide appears to be an improvement over the orange test color of the original modified reagent. Hence, further investigation of the potassium iodide formulation will be made with attention centered on recent findings of B. Fromm, Army Chemical Center, that the modified Dragendorff's reagent is stabilized by certain carbohydrates. Possibly, the presence of some material such as sugars or starch will stabilize the potassium iodide formulation and it will still retain the striking color change.

The color body derived from the reaction of modified Dragendorff's reagent with amines differs from the bismuth triiodide-amine salts previously characterized. Preliminary analysis of a colored product* isolated from the reaction of tri-n-butylamine and the modified Dragendorff's reagent indicated that the specimen did not contain cadmium, but may contain appreciable quantities of chlorine in addition to iodine. Insufficient amounts of sample prevented specific analysis of each of the halogen constituents. Attempts have been made to duplicate the ACC preparation to provide sufficient sample for complete analysis, but none of these have yielded a product suitable for analysis.

Spectroscopic and elemental analysis are also being obtained on the modified Dragendorff's reagent. Both the aqueous solution and the insoluble precipitate are being analysed to determine their composition.

Quinone Oximes

Preliminary examination of quinone monoxime for the detection of V- and G-agents gave encouraging results with benzenesulfonyl chloride but not with EA-1701. The proposed test is based on the fact that arylsulfonates of quinone monoximes readily react with phenols in pyridine solutions to form intensely colored indophenols**. Since mercuric bromide and zinc thiocyanate accelerated the reaction with benzenesulfonyl chloride it seemed desirable to pursue this approach. For this evaluation the quinone oximes shown below were reacted with benzenesulfonyl chloride, benzoyl chloride, and diethylethane phosphonate in solutions of pyridine with and without phenol.

Quinone Monoxime (Sodium salt of p-nitrosophenol)

2-Hydroxyquinone monoxime (Sodium salt of nitrosoresorcinol)

*Submitted by the Army Chemical Center.

**Burmistrov, et al., *Ukrain. Khim. Zhur.*, 22, 323 (1956); cf. C. A. 51, 272 H. (1957).

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2-Methyl-5-isopropylquinone monoxime (Nitrosothymol)

4-Methoxy-1, 2-quinonemonoxime (5-methoxy-2-nitrosophenol)

Since 2-methyl-5-isopropylquinone monoxime showed the most promise in these tests, the effect of the mercuric bromide and zinc thiocyanate salts, as well as potassium cyanide, on its reactions with the same acylating agents was examined. Only benzene-sulfonyl chloride consistently led to intensely colored products; benzoyl chloride was much less effective and diethylethane phosphonate was ineffective. A few spot tests, using 1 per cent EA-1701 in benzene as acylating agent, did not show promise. Apparently, the conversion of quinone oximes to indophenols through the action of acylating agents is limited to strong acylating agents. Hence, the test would not appear to be applicable for the detection of V-agents, since such agents normally behave as weak acylating agents. Further work on this phase of the program will be discontinued unless an attractive modification of the reaction is found.

Detection of V- and G-Agents in the Presence of Decontaminating Agents

Acid Indicators

Congo Red. Of the acid indicators tested for color reactions with EA-1701 in the presence of zinc oxide and decontaminating agents, Congo red appeared to show the most promise. To further examine its possibilities mixtures of Congo red were made with CC-2 (an N-chloro-substituted diphenyl urea) and CC-3^a (a 10 per cent zinc oxide-stabilized CC-2) at levels of 1 per cent and 0.1 per cent Congo red by grinding the powders together in a mortar. A purple color slowly developed in both of the mixtures of Congo red and CC-3, and the 1 per cent mixture of Congo red and CC-2. This purple color appeared to reach a maximum in about 1 week when the samples were stored in the dark at room temperature. Only the 0.1 per cent mixture of Congo red and CC-2 failed to develop this purple color. In fact, the pink color of the mixture appeared to fade, possibly through reaction of the chlorinating agent with the Congo red. In tests with these materials three drops of 1 per cent EA-1701 in ethyl alcohol or benzene were added to 5 to 10 milligrams of the solids contained in small test tubes. The benzene solution of EA-1701 changed the color of the solids to reddish purple similar to the color formed during the storage of the powders containing 1 per cent of Congo red and CC-2 or CC-3. The alcohol solution of V-agent produced a slight purple color in the 1 per cent Congo red-CC-3 mixture, but a yellow color in the 1 per cent Congo red-CC-2 mixture. The solutions of V-agent, however, bleached the pink color of the 0.1 per cent Congo red mixtures. This action required 1 hour in the case of the alcoholic solution and 3 to 4 hours in the case of the benzene solution.

These tests indicate that CC-2 and CC-3 interact with Congo red to destroy its acid-base indicating properties. Also, it appears that enough acid is produced by CC-2 or CC-3 to give a false positive test. Therefore, at this time, it would appear that the

^aSample obtained from A. Oglesby, Army Chemical Center.

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use of acid indicators for the detection of V- or G-agent in the presence of a decontaminating agent does not hold much promise of success.

Oxidation-Reduction Indicators

Tetrazolium Compounds. The promising results of preliminary tests on EA-1701 using reduction indicators such as triphenyltetrazolium chloride or 3,3'-(3,3'-dimethoxy-4,4'-biphenylene)bis [2,5-diphenyltetrazolium chloride] (blue tetrazolium) prompted a literature check on the properties of tetrazolium salts. Apparently, tetrazolium compounds have been extensively studied only in recent years, yet many compounds have been examined, primarily in biological applications. A survey of the field is covered in an excellent review by A. W. Nineham². Through information contained in this survey, 2-(4-iodophenyl)-3-(4-nitrophenyl)-5-phenyltetrazolium chloride²² (III), and 2-(5-methylthiazol-2-yl)-3,5-diphenyltetrazolium bromide²³ (IV) were selected for study because



they are reported to be more stable to light and are reduced to formazans that are more highly colored than those from triphenyltetrazolium chloride. For example, the thiazol-substituted tetrazole is said to be stable to sunlight when deposited on paper from 0.1 per cent solutions containing sodium bicarbonate. Tetrazolium salts normally do not exhibit this stability, and light converts them to the formazans and other highly colored products. Thus, 2,3(2,2'-diphenylene)-5-phenyltetrazolium chloride has been isolated from the photo oxidation products of triphenyltetrazolium chloride.

FUTURE WORK

The development of tests for the detection of V-agents through their phosphonate, amino, and thiol functions will be continued along the lines that have been established

²²Chemical Reviews, 33, 355 (1953).

²³Science, 81, 385 (1950).

²⁴Chem. Ber., 87, 1505 (1954).

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In preceding progress reports. Thus, additional efforts will be made to improve the detector crayons and test papers based on Michler's ketone oxime systems. The search for more stable oxime reagents will be focused primarily on the ortho-isomer of MMK-O and the DIA-MMK-O hybrid illustrated earlier in this report. In the present state of development, quinone monoximes do not appear to hold much promise as reagents for the detection of V- or G-agents, and work on this phase of the program will be discontinued. Evaluation of the modified Dragendorff's reagent system will include analysis of the water soluble and insoluble phases of the reagent as well as complete analysis of the final color body derived from its reaction with a tertiary amine. Attempts will be made to stabilize the modified Dragendorff's reagent containing potassium iodide. Studies on halogenated quinones will be directed toward new and more sensitive reagents.

The use of acid indicators for the detection of V- or G-agents or their degradation products in the presence of decontaminants has not given encouraging results. Consequently, other means of detection will be pursued. At the present time, tetrazolium salts warrant investigation for the detection of V-agents. In exploratory tests with solutions of EA-1701, triphenyltetrazolium chloride and Blue Tetrazolium gave the corresponding red and blue formazans, presumably by reaction through the thiol function of the agent. Among the probable disadvantageous properties of tetrazolium salts, lack of specificity and instability to light would limit their usefulness as reagents for the detection of V-agents. However, Nineham's review on formazans and tetrazolium salts cites several tetrazoles that appear to overcome these deficiencies. Of these 2-(4-iodophenyl)-3-(4-nitrophenyl)-5-phenyl tetrazolium chloride and 2-(5-methylthiazol-2-yl)-3,5-diphenyltetrazolium bromide will be prepared and examined.

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APPENDIX

STUDY OF THE NATURE OF THE REACTION OF HALOGENATED QUINONES WITH AMINES

Reactions of Chloranil

Spectrophotometric investigation of the color development arising from the reaction of 1×10^{-3} molar solutions of triethylamine and chloranil in chloroform has shown that the rate of color formation at room temperature is quite slow. Even at the end of 5 days the color producing reaction was incomplete, although the rate had slowed down appreciably after 24 hours. Doubling the triethylamine concentration (to 2×10^{-3} molar) and keeping the chloranil concentration the same (1×10^{-3} molar) increased the optical density of the solution at the end of 24 hours by a factor of about 1.7. Reduction of the triethylamine concentration of 4×10^{-4} molar at the same chloranil concentration (1×10^{-3} molar) decreased the optical density at the end of 24 hours by a factor of 0.4. Further reduction of the triethylamine concentration to 2×10^{-4} molar and 1×10^{-4} molar resulted in no measurable blue color formation. It is possible that at these lower levels of amine concentration there was sufficient acid formation from the chloranil or chloroform to convert the amine to its hydrochloride salt, thus preventing color formation.

Attempts were made to accelerate the amine-chloranil reaction by the use of mercuric bromide and the sterically hindered bases, 2,6-lutidine and quinaldine. None of these approaches proved successful. In fact, use of mercuric bromide prevented the visible formation of the color body for several hours. Only after a precipitate formed, presumably of mercuric origin was any color formed.

The system dimethyl aniline-chloranil in chloroform was briefly investigated. The color body formed quite rapidly in this case, but required a high concentration of the amine, approximately 1/10 molar, to be measurable.

Preparation of Crayons

MMK-O and EMK-O-Zinc Thiocyanate Crayons

Zinc thiocyanate normally enhances deterioration of Michler's ketone oximes, so that best results were obtained by incorporating these agents into separate powders. By blending the stock powders immediately before pressing into crayons, and eliminating the small amount of water that is normally added as lubricant, pale yellow crayons with good writing qualities were obtained. The composition of a typical MMK-O crayon formulation is shown on page 16:

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Powder A

Michler's ketone oxime (MMK-O)	1.0 part
Chalk powder ^(a)	4.0 parts

Powder B

Zinc thiocyanate	1.0 part
Chalk powder ^(a)	4.0 parts

(a) From ground An-di-cyclic white blackboard crayon made by Stacey and Smith Co., New York, N. Y.

When oxime stabilizers were studied they were incorporated along with the oxime in Powder A. In such cases, that amount of chalk used was diminished in proportion to the amount of stabilizer used, usually 0.2 part. Thus, Powders A and B, were prepared separately by grinding the designated ingredients in a mortar. The stock powders that resulted were intermixed without grinding and were finally pressed into crayons using a 7/16-inch split mold under a pressure of 33,400 pounds per square inch.

Preparation of Reagents

Sodium Salt of Isanitroso-4-Dimethylaminoacetophenone

To a solution of 2 g. of sodium in 30 ml. of absolute alcohol was added with ice-water cooling and swirling 10.3 g. of n-butyl nitrite. To this mixture was added 10 g. of 4-dimethylaminoacetophenone and about 10 ml. more of alcohol. After standing at room temperature for 2 days, the mixture was heated on a steam bath for 1 hour; this treatment changed the appearance of the solid lumps of unreacted ketone and the desired product precipitated as a tan powder. This product was collected by suction filtration, washed thoroughly with ether, and air dried; yield, 8.2 g. (63 per cent) of the sodium salt of 4-dimethylaminoisnitrosoacetophenone.

2-Dimethylaminoacetophenone Oxime

The reaction of N, N-dimethylaniline with acetic acid and phosphorus pentoxide to form 4-dimethylaminoacetophenone* also yielded a lower-boiling yellow oil, b. p. 128° at 16 mm., that corresponded to 2-dimethylaminoacetophenone**. Oximation of the oil was accomplished by reaction with 1 mole excess of free hydroxylamine in alcohol at room temperature for about 20 hours. Concentration of the reaction mixture, cooling, and dilution with petroleum ether-ethyl ether, separated the oxime as white

*Nicham, J. Chem. Soc., 633 (1952).

**Grammaticakis, Compt. Rend. 235, 646 (1952).

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granular crystals. Apparently, preparation of the hydroxylamine solution from hydroxylamine hydrochloride and potassium hydroxide left considerable amounts of hydroxylamine salt in solution, for the ketone product melted over a wide range (84 to 145°C.). The salt was liberated by dissolving the crude oxime in water, making the solution strongly alkaline with sodium carbonate, and removing the oil that eventually solidified upon cooling. Recrystallization of the oxime by addition of water to a cooled ethyl alcohol solution of the crude product separated white needles, m.p. 84-84.5°C. (Lit., m.p. 84°C.).

N-Acetyl Auramine

As indicated in the Tenth Monthly Progress Report, preparation of N-acetyl auramine following the procedure of Semper* gave yellow crystalline material that reacted as reported, but melted at 115 to 117°C. instead of the reported value of 221°C. Elemental analysis of the experimental specimen was made and the values obtained for carbon hydrogen and nitrogen are in good agreement with those calculated for N-acetyl auramine. Hence, it was concluded that either the melting point reported by Semper is in error, or the two compounds are structural isomers.

Anal. Calc'd. for $C_{19}H_{23}N_3O$: C, 73.76; H, 7.50; N, 13.59.

Found: C, 73.43; H, 7.22; N, 13.60.

*Ann., 351, 234 (1911).

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